Bell inequality in chiral liquids

Sara Restuccia[a], Graham M. Gibson[a], Leroy Cronin[b], and Miles J. Padgett[a]

[a]School of Physics and Astronomy, University of Glasgow, Glasgow G12 8QQ, United Kingdom
[b]WestCHEM, Department of Chemistry, University of Glasgow, Glasgow G12 8QQ, United Kingdom

ABSTRACT

Optical activity is a macroscopic property of chiral molecules which manifests as a rotation of the plane of linear polarization when light passes through a sample. We have developed a compact Bell inequality experiment for quantum probing of chiral liquids, based on polarization measurements. In particular, we show that we can use a Bell-type inequality configuration to measure the optical activity of D-Limonene, a chiral molecule which is a major component in the oil of citrus fruit peels.

Keywords: Quantum Polarimetry, chiral solutions, entanglement

1. INTRODUCTION

Chiral objects are materials that lack mirror symmetry i.e., a chiral object cannot be superposed onto its mirror image. A keen interest exists in the measurement of the optical activity of chiral materials as chirality plays an important role in a variety of fields, including physics, biology, chemistry, and materials science.[1–4] One of the methods by which chirality can be tested is through the interaction of a linearly polarised beam of light. This interaction results in a measurable rotation in the polarisation of the transmitted light beam. By measuring the rotation of the linear polarization of light following its propagation through a chiral medium of known path length, the optical activity of the material can be determined. Knowing the chirality of a molecule is essential in fields like the pharmaceutical industry as having the wrong chiral molecule can render a drug ineffective or toxic.[5] It is important to note that in a classical detection scheme used to measure the optical activity of a chiral medium, the precision of the measurement increases with the intensity of the light used.[4] However, this becomes problematic in situations where the intensity of the incident light might cause damage to the sample (for example, when measuring the chirality of samples containing proteins[3]) or triggers unwanted chemical reactions (such as in the probing of molecular material). Alternatively, a quantum detection scheme uses spontaneous parametric down-converted (SPDC) photons as the light source. The system presented here is a compact and simple Bell-type inequality configuration that is capable of measuring the chirality of a molecule in solution by performing polarisation measurements at the single photon regime.

2. EXPERIMENTAL SET-UP

In this work, we constructed a compact Bell-type inequality set-up in which a chiral solution (D-Limonene, in this case) can be easily inserted and its optical activity measured. In order to achieve this, careful consideration was taken in choosing both the source of our entangled down-converted photons and the detection system employed to measure photons.

For the generation of the entangled photons, a type-I SPDC source was chosen analogous to that first described by Kwiat *et al.* in their 1999 paper.[6] In this set-up, a two-crystal geometry is used to obtain polarization entangled photons. Two identical BBO crystals, each cut for type-I phase matching, are 'sandwiched' together with their optical axis orientated perpendicular to one other. More specifically, the two identical crystals are orientated at 90° to each other with respect to the pump propagation direction. In particular, the first crystal's optical axis is arranged parallel to the vertical plane of the pump propagation direction while the second crystal axis lies on the horizontal plane. In this set up, if the pump beam is vertically polarised, the SPDC process

Adress all correspondence to Sara Restuccia, E-mail: Sara.Restuccia@glasgow.ac.uk

will only occur in the first crystal and the outgoing photons will be generated in a horizontally polarized, down conversion light cone. Similarly, if the pump beam is horizontally polarised, the SPDC process will only occur in the second crystal and the output cone will be vertically polarised. To generate polarization entangled photons, the pump beam needs to be orientated at 45° with respect to the optical axis of each of the two 'sandwiched' crystals. This will allow the SPDC process to be equally likely to occur in either crystal, with the two possible processes occurring as a coherent superposition as long as the emitted spatial modes of the photon pairs are indistinguishable for the two crystals i.e., the cones overlap or are too close together to determine from which one the photon originated (Figure 1).

The advantage of using a two-crystal geometry (cut for type-I phase-matching) over a single crystal (cut for type-II phase-matching), is that the signal and idler beams overlap in the far-field of the crystal. As a consequence, for the two-crystal geometry the full downconversion beam or ring is polarization entangled as opposed to only two special directions or the overlap of the spatially separated rings, as is the case for type-II phase-matching.[7] This makes the set-up convenient for probing cells containing a chiral solution as it allows for additional down converted photons to interact with the solution before being collected for detection.

The choice of detectors for the system plays an important role. The collecting aperture of the detectors have to be a large enough aperture to collect photons generated by the SPDC source which, as mentioned above, are not generated in a strict direction but can be found at any point around the overlapping cones. It is also important to mention that although choosing a detector with a bigger aperture will inevitably collect more noise, it will also allow the system to be less sensitive to small misalignment errors. These errors could arise from swapping the sample cell in and out of the system. In particular, choosing a wide aperture bucket detector will allow for the measurement of photons with a wider range of acceptance angles making the system quicker and simpler to use.

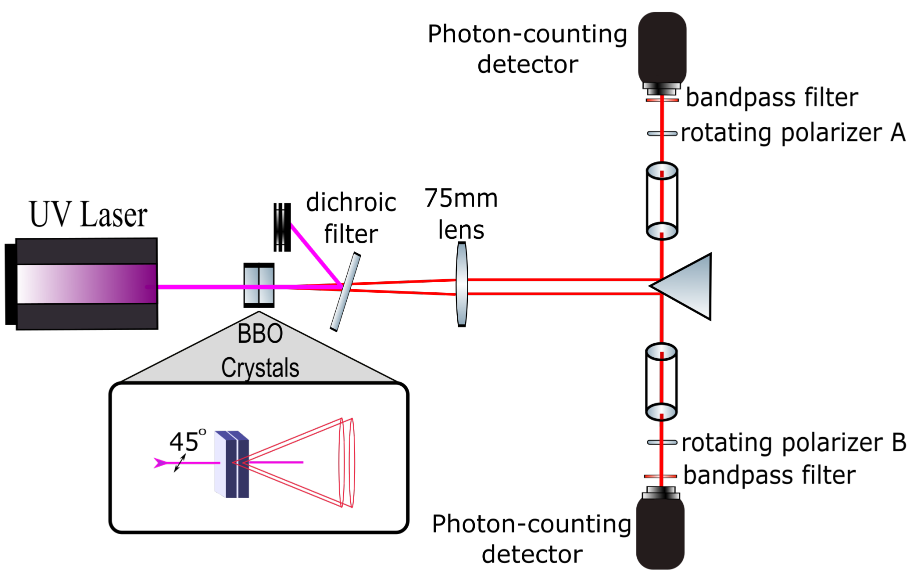

Figure 1. Experimental set-up. A type-I two-crystal geometry SPDC source is employed to generating a polarized entangled downconversion beam. The signal and idler components of the rings are separated in the far field by a knife-edge prism into two arms each containing: a sample cell holder, a motorised polarizer, a 10 nm bandpass filter and a detector. The detectors are connected to a coincidence counter which allows for the measurement of coincidences as a function of the polarizer angle.

A schematic of the set-up designed for this experiment is shown in Figure 1. A 355 nm laser (JDSU, xCyte

CY-355-150) generates our pump beam. This beam is attenuated using a neutral density filter of optical density 0.6 and is then passed through a half wave-plate to orientate the UV photons at 45° with respect to the optical axis of each of the two downconversion crystals that comprise the SPDC source. The resulting UV photons interact with a system comprising of a 'sandwich' of two paired 1 mm β-barium borate (BBO) nonlinear crystals, cut for type-I phase-matching and degenerate downconversion. A dichroic mirror is used to remove the residual UV pump, leaving only the entangled photon-pairs at a degenerate wavelength of 710 nm in the system. The signal and idler beams are then separated from one other using a knife-edge prism in the far field of the down-conversion crystals, directing them into two separate identical arms of the system. Each of the two arms are composed of a holder for inserting a sample cell, a polarizer set on a motorized rotation stage, a 10 nm bandpass filter and finally a photon counting detector. The outputs of the photon counting detectors in each arm are connected to a coincidence counter which allows for the measurement of coincidences as a function of the polarizer angle. As mentioned above, there is a distinct advantage in choosing a detector with a wide sensitive area. In our system we therefore used a H7421 series photon counting head which is composed of a photomultiplier tube with a 5 mm diameter sensitive area. It is important to mention that having a larger sensitive area for the detectors will inevitably increase the number of background photon events picked up by the detectors which can give rise to random coincidence events (i.e. 'accidentals'). As non classical states are independent of each other, they follow a Poissonian distribution.[8] This is done to diminish the number of 'accidental' coincidence counts by simply limiting the coincidence rates measured solely to those in which the Poissonian peak appears. In our experiment, we set a coincidence window of 1.8 ns. To furthermore limit the accidentals in the system, a 10 nm bandpass filter (centered at 710 nm) is mounted in front of each detector. As the experimental setup is resilient to the effects of small misalignments, it allows for the use of low cost, more compact optomechanics. In particular, our system uses both 3D printed rotating polarizer mounts and 3D printed cell sample cell holders.

3. PRELIMINARY RESULTS

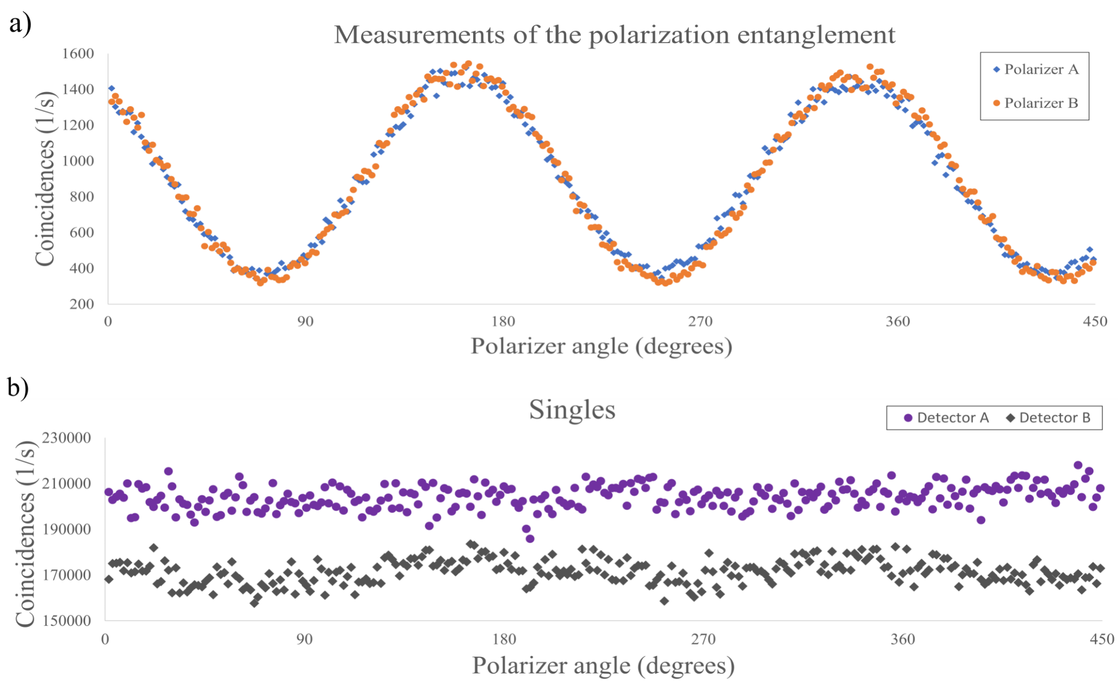

Figure 2. a) Correlation measurements as a function of polarizer angle. Coincidences are measured by rotating the polarizer in one of the experiment arms while keeping the other fixed. The measurement was taken for both polarizer A and polarizer B. b) Number of single photons recorded by detector A and detector B during the coincidence window in which the coincidence as a function of the rotation of polarizer A are calculated.

As we are interested in constructing a Bell set up to probe chiral solutions, the first test is to demonstrate the efficiency of the system. Hence, the photon number and coincidence counts were measured as a function of rotation of one of the polarizers in the system. In particular, the first measurement was performed by rotating the polarizer on Arm A while keeping the polarizer on Arm B fixed (Figures 2a and 2b). As noted in Figure 2a, rotating the coincidence recorded display the expected quantum-mechanical correlations while the individual single photons remain constant (Figure 2b). Having unchanged values of singles as a function of polarization angle implies that the photons collected are individually nearly unpolarized. Hence, the measurement of the sinusoidal coincidence fringes is due to the presence of polarized entangled photons in our system. The measurement is repeated by rotating the polarizer in Arm B with the polarizer in Arm A fixed (Figure 2). The resulting sinusoidal curve measured was in good agreement with the previous measurement in which the angle difference between the two polarizers was determined by rotating polarizer A. This is in agreement with what expected for a quantum Bell measurement where the rate of coincidences are determined by the relative angle difference between the polarizers, irrespective of which polarizer is rotated.

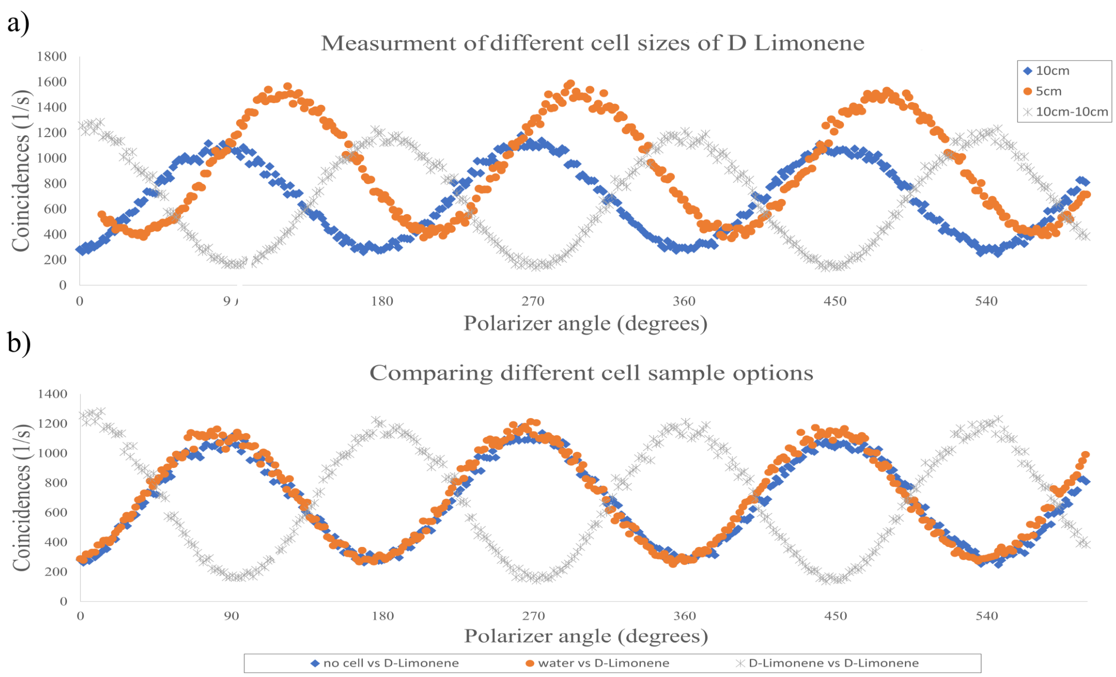

Figure 3. Figure a) illustrates the correlation measurements as a function of polarization angle when cells of different length are inserted in the system. In particular, it shows the shift in the Bell curve when a 5cm cell containing D-Limonene, is inserted in one of the arms; a 10 cm cell is inserted in one of the arms; and a 10cm cell is inserted in both of the arms. Figure b) compares the Bell correlations when only a 10cm cell containing D-Limonene is present in one of the arms; when a 10cm cell containing D-Limonene is present in both arms; and when a 10cm cell containing distilled water in present in one arm while the second arm contains a 10cm cell of D-Limonene.

Having demonstrated the capability of the system to generate polarized entangled photons, some preliminary data was taken to probe the system's ability to measure the optical activity of a chiral solution i.e., measure a rotation in the linear polarization of light after its interaction with the chiral medium. In particular, three consecutive correlation measurements as a function of polarizer angle were taken and the resulting sinusoidal fringes compared. The first measurement was taken with no sample present in Arm A and a 5 cm cell containing D-Limonene present in Arm B. Similarly, a second measurement was taken with no sample present in Arm A and a 10 cm cell of D-Limonene in Arm B. Finally, a third measurement was performed with a 10 cm cell of solution placed in both arms. Having a cell of chiral solution in both arms is equivalent to having no cell in the optical system. As seen in figure 3a, the chiral solution in one of the arms of the system produces a phase shift in the sinusoidal coincidence fringes. In particular, the 10 cm cell produces a shift of approximately 100°while the 5 cm cell of 66°. It is important to state that the results are preliminary and more accurate optomechanics are currently being designed for the system to generate a precise measurement. Nevertheless, the preliminary results are consistent with both the expectation of a double shift for a 10 cm cell compared to the 5 cm cell and the known chirality of D-limonene of 123°.[9]

Finally, a comparison was also made between the Bell correlations when only a 10 cm cell containing D-Limonene is present in one of the arms and when a 10 cm cell containing distilled water (a non-chiral solution) is present in one arm while the second arm contains a 10 cm cell of of D-Limonene. As distilled water has no chiral properties, no optical rotation is expected for the photons that interact with it. As seen from figure 3b, placing a non-chiral solution in the system is equivalent to having no solution.

4. CONCLUSIONS AND FUTURE WORK

The preliminary data collected for the compact Bell inequality system we have designed show that the set-up is capable of generating polarization entanglement photons and that these photons can potentially be used for precise measurement of the chirality of molecules at the single photon regime. The use of a Bell-type inequality configuration will also allow measurements to be performed to assess the degree of entanglement in the system.[10] In particular, a Clauser-Horne-Shimony-Holt (CHSH) Bell inequality is often used and a Bell inequality is deemed to be violated should $|S| \leq 2$.[11] As has been shown, the presence of a chiral solution in one of the arm of the system generates a controlled optical rotation of the polarization of the photon that have interacted with it. Bell measurements on polarisation entangled photon-pairs entail measuring the rate of coincidences as a function of the relative angles of the pair of polarizers placed on each arm. As seen from the preliminary data, the presence of a chiral solution of given length will shift the Bell curve i.e., the rate of coincidence will no longer be minimum when the polarizers are the same and maximum when the polarizers are orthogonal to each other. More importantly, the original Bell curve can be regained by inserting a solution with the same optical rotation in the opposite arm. Measuring the degree of entanglement when using the chiral solution could potentially be used to hide and recover the entanglement in the system. To control, and potentially hide and recover entanglement for encryption of a signal could be of great importance in the development of quantum communications systems. The use of entangled light sources for quantum communications is the subject of great interest as such schemes offer secure means of communication.

ACKNOWLEDGMENTS

We acknowledge financial support from the EPSRC (UK Grant No. EP/S019472/1) and from The Leverhulme Trust.

REFERENCES

[1] Lough, W. J. and Wainer, I. W., [*Chirality in natural and applied science*], Blackwell Science (2002).

[2] Tischler, N., Krenn, M., Fickler, R., Vidal, X., Zeilinger, A., and Molina-Terriza, G., "Quantum optical rotatory dispersion," *Science Advances* **2**(10), e1601306 (2016).

[3] Cimini, V., Mellini, M., Rampioni, G., Sbroscia, M., Leoni, L., Barbieri, M., and Gianani, I., "Adaptive tracking of enzymatic reactions with quantum light," *Optics Express* **27**, 35245–35256 (nov 2019).

[4] Yoon, S.-J., Lee, J.-S., Rockstuhl, C., Lee, C., and Lee, K.-G., "Experimental quantum polarimetry using heralded single photons," *Metrologia* **57**, 45008 (jun 2020).

[5] Hutt, A. J. and Tan, S. C., "Drug Chirality and its Clinical Significance," *Drugs* **52**(5), 1–12 (1996).

[6] Kwiat, P. G., Waks, E., White, A. G., Appelbaum, I., and Eberhard, P. H., "Ultrabright source of polarization-entangled photons," *Physical Review A* **60**, R773–R776 (Aug 1999).

[7] Kwiat, P. G., Mattle, K., Weinfurter, H., Zeilinger, A., Sergienko, A. V., and Shih, Y., "New High-Intensity Source of Polarization-Entangled Photon Pairs," *Physical Review Letters* **75**, 4337–4341 (Dec 1995).

[8] Couteau, C., "Spontaneous parametric down-conversion," *Contemporary Physics* **59**(3), 291–304 (2018).

[9] Lide, D. R. and A., M. G. W., [*Handbook of data on common organic compounds*], CRC Press (1995).

[10] Bell, J. S., "On the Einstein Podolsky Rosen paradox," *Physics Physique Fizika* **1**, 195–200 (Nov 1964).

[11] Clauser, J. F., Horne, M. A., Shimony, A., and Holt, R. A., "Proposed experiment to test local hidden-variable theories," *Physical Review Letters* **23**, 880–884 (Oct 1969).